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In re application of:

Date: January 14, 2004

Alan W. Peters et al.

Art Unit: 1764

Serial No.: 10/022,102

Examiner: Yildirim, Bekir L.

Filed: December 18, 2001

Docket No.: W9003-04

For: REDUCED NO_x COMBUSTION PROMOTER FOR USE IN FCC PROCESSES

DECLARATION UNDER 37 CFR 1.132

I, George Yaluris, hereby declare as follows

1. I received my Ph.D. in chemical engineering from the University of Wisconsin-Madison in 1995.
2. From 1995 to present I have been employed by and continue to work for W. R. Grace & Co.-Conn.
3. During my employment at W. R. Grace & Co.-Conn., I have held the position of Research Engineer, Senior Research Engineer, Principal Engineer, and currently Marketing Specialist.
4. I am a named author in at least 14 peer reviewed publications, including two peer reviewed publications on the origin and control of NO_x in a Fluid Catalytic Cracking (FCC) unit regenerator, as well as on the development of new methods for testing in the laboratory NO_x reducing catalytic compositions.
5. I am a named author in at least 11 other publications in trade journals and other publications, including two publications on NO_x chemistry and control in the FCC unit regenerator.
6. I am a named inventor in one U.S. patent issued, and in four U.S. patent applications currently pending with the PTO.
7. I worked closely with Dr. Peters, now deceased, one of the inventors of the present invention, and I am familiar with the contents of the above referenced patent application.

8. In an effort to demonstrate that the composition claimed in the subject application is effective for controlling CO emissions while simultaneously reducing NOx emissions when the alkaline earth oxide content of the composition is less than 10%, three samples, A, B and C, having a general composition as claimed in the subject application and a specific composition as shown in Table 1, were tested under my direction and control for NOx reduction and CO emissions control:

Table 1
Composition of samples tested (dry basis)

		Sample A	Sample B	Sample C
SiO₂	: wt%	6.5	6.642	6.437
Al₂O₃	: wt%	64.638	61.47	59.551
CeO₂	: wt%	22.022	20.73	20.369
Na₂O	: wt%	0.263	0.149	0.395
MgO	: wt%	7.732	8.2	15.989
Pd	: ppm	915	1077	915

9. To demonstrate the ability of Samples A - C to reduce NO emissions while controlling CO emissions, the Samples were tested both in our Regenerator Test Unit (RTU) and an FCC pilot plant, the Davison Circulating Riser (DCR). The description of the RTU has been published in G. Yaluris and A. W. Peters "Studying the Chemistry of the FCCU Regenerator Under Realistic conditions," *in* Designing Transportation Fuels for a Cleaner Environment, J.G. Reynolds and M.R. Khan, eds., p. 151, Taylor & Francis, 1999, ISBN 1-56032-813-4. The description of the DCR has been published in the following papers: G. W. Young, G. D. Weatherbee, and S. W. Davey, "Simulating Commercial FCCU yields with the Davison Circulating Riser (DCR) pilot plant unit," National Petroleum Refiners Association (NPRA) Paper AM88-52; G. W. Young, "Realistic Assessment of FCC Catalyst Performance in the Laboratory," *in* Fluid Catalytic Cracking: Science and Technology, J. S. Magee and M. M. Mitchell, Jr. Eds., Studies in Surface Science and Catalysis Volume 76, p. 257, Elsevier Science Publishers B.V., Amsterdam 1993, ISBN 0-444-89037-8. Testing in the RTU measures the ability of a composition to control CO emissions, while testing in the DCR measures the ability of the composition to control NO emissions.
10. The CO oxidation activity of Samples A-C was tested in the RTU along with a commercially available sample of Pt-based combustion promoter, CP-3[®]

The cracking catalyst was first deactivated for 4 h at 1500 °F in a fluidized bed reactor with 100% steam before being coked. The properties of this catalyst are shown in Table 2.

Table 2
Properties of coked catalyst used in RTU tests.

CHEMICAL ANALYSES:		
SiO₂-Diff		50.7
Al₂O₃	: Wt. %	45.003
RE₂O₃	: Wt. %	1.99
Na₂O	: Wt. %	0.243
SO₄	: Wt. %	0.37
Fe₂O₃	: Wt. %	0.5
TiO₂	: Wt. %	0.7
MgO	: Wt. %	0.054
P₂O₅	: Wt. %	0.120
CaO	: Wt. %	0.085
Ni	: ppm	49
V	: WT. %	0.007
Coke	wt %	1.522

Samples A - C and the CP-3[®] were calcined for 2 h at 1100 °F and then mixed with the coked cracking catalyst at 0.2% additive level (CP-3[®] was mixed at 0.1% additive level). The blend was then fed into the RTU reactor operating at 700 or 710 °C and the coke was burnt with a gas mixture of air and nitrogen so that there was 1.1% excess O₂ in the RTU reactor effluent.

The CO measured at the reactor effluent is a measure of the CO combustion activity of the sample tested. The lower the measured CO the higher the combustion activity of the additive.

11. Samples A – C were tested in the DCR along with CP-3[®]. The DCR was started up by charging the unit with approximately 1900 g of equilibrium catalyst, designated as EQ98-43. The properties of this catalyst are summarized in Table 3. For the purposes of this test, a commercial FCC feed was used having the properties shown in Table 4.

Table 3
Properties of feed FCC equilibrium catalyst used in the DCR tests

		EQ98-43
CHEMICAL ANALYSES:		
SiO₂	: wt. %	50.91
Al₂O₃	: wt. %	45.5
RE₂O₃	: wt. %	0.37
Na₂O	: wt. %	0.37
Ni	: ppm	681
V	: ppm	1160
PHYSICAL ANALYSES:		
Thermal: 2/1100 or 3/1000 °F		
ABD	: cc/g	0.72
DI		7
APS	microns	60
SA	: m ² /g	195
Zeolite	: m ² /g	141
Unit Cell		24.24
MA	: wt. %	73

Table 4
Properties of feed used in the DCR tests.

API Gravity @60 °F	22.8
Sulfur, wt. %	0.23
Total Nitrogen, wt. %	0.19
Basic Nitrogen, wt. %	0.083
Conradson Carbon, wt. %	1.44
K Factor	11.65
Refractive Index	1.511104
Simulated Distillation, vol. %, °F	
5	464
20	651
40	764
60	854
80	963
95	1207
FBP	1365

The DCR was operated with 1% excess O₂ in the regenerator, and with the regenerator operating at 705 °C. After the unit stabilized, the baseline SO₂ and NO emissions data were collected using an on-line Lear-Siegler SO₂/NO Analyzer (SM8100A). Subsequently, 100 g of catalyst were injected into the DCR consisting of 10 g of Samples A, B or C, or 5 g of CP-3[®], and the balance equilibrium catalyst.

12. It is well known that conventional combustion promoters commonly used to control CO in the FCC unit regenerator can cause NO emissions to increase. The results of my tests, shown in Table 5, clearly demonstrate that all Samples tested (A - C and CP-3[®]) are active and effective for controlling CO emissions from an FCCU regenerator, and that CP-3[®] increases NO emissions as expected. However, only samples A and B, containing less than 10% MgO were able to simultaneously control CO emissions and achieve NO emissions that were lower than those of the conventional Pt-based promoter (CP-3[®]) used as a reference. When the MgO content of the composition exceeded 10%, the composition was still able to control CO emissions, but the NO emissions increased compared to those of the reference conventional CO combustion promoter.

Table 5
Results of the RTU and DCR tests of Samples A - C and CP-3[®].

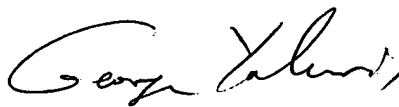
Additive	DCR			RTU Level (%)	CO RTU 700 °C (ppm)	CO RTU 710 °C (ppm)
	Level (%)	Flue Gas (l/h)	Normalized NO* (ppm)			
Cracking Catalyst CP-3	0	1060.7	22	0	4923	2198
	0.25	1058	257	0.1	718	535
Cracking Catalyst Sample A	0	1138	53	0	4923	2198
	0.5	1149	214	0.2	1317	960
Cracking Catalyst Sample B	0	1152	47	0	4923	2198
	0.5	1161	237	0.2	1038	751
Cracking Catalyst Sample C	0	1150	62	0	4923	2198
	0.5	1158	268	0.2	1165	826

* NO normalized to 1150 l/h flue gas flow

13. I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

01/15/04

Date



George Yaluris, PhD